Foundations for Cooperating with Control Noise in the Manipulation of Quantum Dynamics

Feng Shuang,[1] Herschel Rabitz,[1] and Mark Dykman[2]

[1] *Department of Chemistry, Princeton University, Princeton, New Jersey 08544*

[2] *Department of Physics and Astronomy, Michigan State University, East Lansing, Michigan 48824*

(Dated: February 1, 2008)

Abstract

This paper develops the theoretical foundations for the ability of a control field to cooperate with noise in the manipulation of quantum dynamics. The noise enters as run-to-run variations in the control amplitudes, phases and frequencies with the observation being an ensemble average over many runs as is commonly done in the laboratory. Weak field perturbation theory is developed to show that noise in the amplitude and frequency components of the control field can enhance the process of population transfer in a multilevel ladder system. The analytical results in this paper support the point that under suitable conditions an optimal field can cooperate with noise to improve the control outcome.

I. INTRODUCTION

Control of quantum processes is actively being pursued theoretically [1, 2] and experimentally[3, 4]. In practice, control field noise and environmental interactions inevitably are involved. The present paper considers the influence of field noise upon the controlled dynamics with the noise described by shot-to-shot pulse variations, as in typical signal averaged experiments. Recent studies considered several aspects of the influence of laser noise[5–11], and this work aims to further explore the issue. The interaction between the noise and field driven dynamics is generally a highly nonlinear process. The impact of the noise can either be constructive[12–15] or destructive[10] in the manipulation of quantum dynamics as well as either improve[16, 17] or reduce[18] the convergence rate of control search efforts. These disparate behaviors make it difficult to precisely identify the role of noise under various circumstances, but the many successful experiments at least support the point that some noise can be tolerated[19–24]. Operating under closed-loop[25] in the laboratory will naturally deal with noise as best as possible. A theoretical analysis[9] on the impact of field noise upon optimal control indicated that an inherent degree of robustness can be anticipated by virtue of the controlled observable expectation values being bilinear in the evolution operator and its adjoint. Some simulations of closed-loop experiments also show that robust control is possible and that properly designed control fields can fight against noise[8, 11, 16, 17].

Recent numerical simulations[11] of closed-loop control in a model system showed that under suitable circumstance the control field could cooperate with the presence of noise to more efficiently reach the target state. The cooperation was remarkable when seeking a modest yield (e.g., $\sim 10\%$) in the target state. In this case the noise or the deterministic field acting alone would each produced a small yield, but the two acting together cooperatively produced a much larger yield. The latter numerical simulations did not reveal the underlying physical origin of the cooperative effect, and the present paper will analyze the controlled dynamics of a multistate system in several limiting cases to explain the prior findings. The cooperative behavior also has foundation in analogous stochastic resonance phenomena[26–28] and fluctuation control[29, 30].

Section II presents a general control model of population transfer in multilevel systems. The noise is modeled by run-to-run variations in the control amplitudes, phases and frequencies with the observation being an ensemble average over many runs as is commonly done in the laboratory. The goal of the control is to maximally populate a highly excited state. In Sec. III we develop a weak field perturbation theory to provide an analytical solution to the outcome of the controlled dynamics. We obtain the noise-average yield from applying the control field in Sec. IV. It is shown that variation of the field phases from pulse-to-pulse plays no role in the dynamics but strong cooperation between the deterministic portion of the field and noise, in both the amplitude and frequency, is possible. Finally, we draw some

conclusions and discuss general multistate systems in Sec. V.

II. THE MODEL SYSTEM

The effects of field noise on controlled quantum dynamics will be explored in the context of population transfer in multilevel systems characterized by the Hamiltonian H,

$$H = H_0 - \mu E(t), \tag{1a}$$

$$H_0 = \sum_n \varepsilon_n |n\rangle \langle n| , \tag{1b}$$

where $|n\rangle$ is an eigenstate of H_0 with the associated energy ε_n in the absence of radiation, and μ is the dipole operator, $\mu = \sum_{n,n'} \mu_{nn'} |n\rangle \langle n'|$. The control field $E(t)$ has the form which may be implemented in the laboratory[31],

$$E(t) = 2s(t) \sum_{l=1}^{M} A_l \cos(\omega_l t + \theta_l) , \tag{2}$$

where $\{\omega_l\}$ are the frequencies of the radiation, and $s(t)$ is the pulse envelope function. The controls are the amplitudes $\{A_l\}$ and phases $\{\theta_l\}$.

Noise in the laboratory could take on various forms and arise from a number of sources[10]. In keeping with laboratory practice, the achieved control will be measured as an ensemble average over the outcome of many noise contaminated control fields, and the noise is modeled as shot-to-shot uncertainties in the amplitudes A_l, phases θ_l and frequencies

ω_l in Eq. (2)[8, 11]:

$$A_l = A_l^0 + \tilde{x}_l, \tag{3a}$$

$$\theta_l = \theta_l^0 + \tilde{y}_l, \tag{3b}$$

$$\omega_l = \omega_l^0 + \tilde{z}_l, \tag{3c}$$

with $\langle \tilde{x}_l \rangle = \langle \tilde{y}_l \rangle = \langle \tilde{z}_l \rangle = 0$. The shot-to-shot field noises are captured in terms of zero-mean uncertainties $\vec{x} = \{\tilde{x}_l\}$, $\vec{y} = \{\tilde{y}_l\}$, and $\vec{z} = \{\tilde{z}_l\}$. For simplicity we assume that the different noise components are independent with distributions $\rho_l^{(A)}(\tilde{x}_l)$, $\rho_l^{(\theta)}(\tilde{y}_l)$ and $\rho_l^{(\omega)}(\tilde{z}_l)$, respectively. In practice the field noise in $E(t)$ may have complex structures and origins. Besides the uncertainties in the control field amplitudes $\{A_l\}$ and phases $\{\theta_l\}$, a potential source of additional uncertainties is in the frequencies $\{\omega_l\}$ due to laser frequency jitter from variations of the refractive index[32], or other sources[33]. The flexible treatment of random variations in $\{A_l\}$, $\{\theta_l\}$ and $\{\omega_l\}$ is meant to represent all of these various possibilities. The net outcome of the control experiments is the average,

$$\bar{O}\left[E(t), \vec{\gamma}\right] = \left(\prod_l \int_{-\gamma_l}^{\gamma_l} \rho_l^{(A)}(\tilde{x}_l)\, d\tilde{x}_l \int_{-\gamma_l'}^{\gamma_l'} \rho_l^{(\theta)}(\tilde{y}_l)\, d\tilde{y}_l \int_{-\gamma_l''}^{\gamma_l''} \rho_l^{(\omega)}(\tilde{z}_l)\, d\tilde{z}_l \right) O\left[E(t, \vec{x}, \vec{y}, \vec{z})\right], \tag{4}$$

where $O\left[E(t, \vec{x}, \vec{y}, \vec{z})\right]$ is the control yield

$$O\left[E(t, \vec{x}, \vec{y}, \vec{z})\right] = \left| \langle \Psi_f | \psi\left[E(t, \vec{x}, \vec{y}, \vec{z}), T\right] \rangle \right|^2, \tag{5}$$

produced by the field $E(t, \vec{x}, \vec{y}, \vec{z})$ in Eq. (2) using the amplitudes, phases and frequencies in Eq. (3). The target state is $|\Psi_f\rangle$, and $|\psi\left[E(t, \vec{x}, \vec{y}, \vec{z}), T\right]\rangle$ is the state of the field-driven

system at the final time T, which is a functional over time of $E(t, \vec{x}, \vec{y}, \vec{z})$, $t \leq T$. In what follows we assume $E(t) \to 0$ for $t \to \pm\infty$.

The objective function to be minimized with respect to $\{A_l^0\}$ and $\{\theta_l^0\}$ in the presence of noise has the form

$$J = \left|\bar{O}\left[E(t), \vec{\gamma}\right] - O_T\right|^2 + \alpha F_0, \tag{6a}$$

$$F_0 = \sum_l \left(A_l^0\right)^2, \tag{6b}$$

where O_T is the target value, and F_0 is the fluence of the control field whose contribution is weighted by the constant, $\alpha > 0$.

III. WEAK FIELD PERTURBATION THEORY

To illustrate the principle of how the deterministic portion of the control field can cooperate with the noise, we consider the excitation along a ladder (or chain) of nondegenerate transitions and energy levels with each linked only to its nearest neighbors. One could analogously think of the system as a nonlinear oscillator[34–36] or a spin with $S > 1$ and nonequidistant energy levels. The transition elements are taken to have the form

$$\mu_{nn'} = \mu_n \delta_{n'+1,n} + \mu_{n'} \delta_{n',n+1}. \tag{7}$$

The $N+1$ level system consists of an initially occupied ground state $|0\rangle$ at $t \to -\infty$, $N-1$ intermediate states $|n\rangle$, $n = 1, 2, \cdots, N-1$, and a final target state $|N\rangle$. The states are

coupled with an external laser pulse having the nominal form of Eq. (2). The wave function is expanded in the form

$$\psi(t) = \sum_{n=0}^{N} C_n\,(t)\,|n\rangle e^{-i\varepsilon_n t}. \tag{8}$$

The initial condition at $t \to -\infty$ specifies that $C_0 = 1$ and $C_n = 0$ for $0 < n \leq N$. The goal is to maximize $|C_N\,(t)|^2$ with $|\psi_f\rangle = |N\rangle$ for $t \to \infty$, when the field is zero. The result of perturbation theory for C_N to the lowest order in the control field $E\,(t)$ is

$$C_N = i^N \prod_{k=1}^{N} \mu_k \int_{-\infty}^{\infty} dt_N E(t_N) e^{i\bar{\omega}_N t_N} \int_{-\infty}^{t_N} dt_{N-1} E(t_{N-1}) e^{i\bar{\omega}_{N-1} t_{N-1}} \ldots \int_{-\infty}^{t_2} dt_1 E(t_1) e^{i\bar{\omega}_1 t_1}, \tag{9}$$

where

$$\bar{\omega}_n = \varepsilon_n - \varepsilon_{n-1},\ n = 1, \cdots, N, \tag{10}$$

are the transition frequencies. Utilizing the Fourier transform of the field

$$E\,(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} f\,(\Omega)\, e^{-i\Omega t} d\Omega, \tag{11a}$$

$$f\,(\Omega) = \int_{-\infty}^{\infty} E\,(t)\, e^{i\Omega t} dt \tag{11b}$$

in Eq. (9) produces

$$C_N = (\frac{i}{2\pi})^N \prod_{k=1}^{N} \mu_k \int d\Omega_1 \cdots d\Omega_N f(\Omega_1) \cdots f\,(\Omega_N)$$

$$\int_{-\infty}^{\infty} dt_N e^{i(\bar{\omega}_N - \Omega_N) t_N} \int_{-\infty}^{t_N} dt_{N-1} e^{i(\bar{\omega}_{N-1} - \Omega_{N-1}) t_{N-1}} \ldots \int_{-\infty}^{t_2} dt_1 e^{i(\bar{\omega}_1 - \Omega_1) t_1}. \tag{12}$$

In the above equation, the definite integrals over the frequencies Ω_k run from $-\infty$ to ∞. To calculate the time integral in Eq. (12), we introduce a small imaginary part $\epsilon_k \to +0$ in the

frequencies,

$$\Omega_k \to \Omega_k + i\varepsilon_k, k = 1, \cdots, N-1. \tag{13}$$

It is seen from Eq. (9) that such a change will not affect the result, since $E(t) \to 0$ for $t \to -\infty$; we assume that $E(t)$ decays at least exponentially for $t \to \pm\infty$, which is physically reasonable. Evaluating the integrals in Eq. (12) with respect to t_1, t_2, $\cdots$, t_{N-1} yields

$$\begin{aligned} C_N = (\frac{i}{2\pi})^N \prod_{k=1}^{N} \mu_k \int d\Omega_1 \cdots d\Omega_N f(\Omega_1) \cdots f(\Omega_N) \\ \int_{-\infty}^{\infty} dt_N e^{-i(\Omega_N - \bar{\omega}_N)t_N} \frac{\exp\left[-i\sum_{q=1}^{N-1}(\Omega_q - \bar{\omega}_q)t_N\right]}{\prod_{j=1}^{N-1}\left[-i\sum_{p=1}^{j}(\Omega_p - \bar{\omega}_p)\right]}, \end{aligned} \tag{14}$$

where all Ω_k, $k = 1, \cdots, N-1$, carry a hidden small positive imaginary part as Eq. (13). Integrating Eq. (14) again with respect to t_N and Ω_N produces C_N in the frequency representation,

$$\begin{aligned} C_N &= \frac{i\prod_{k=1}^{N}\mu_k}{(-2\pi)^{N-1}} \int \prod_{j=1}^{N-1} \frac{f(\Omega_j)d\Omega_j}{\sum_{p=1}^{j}(\Omega_p - \bar{\omega}_p)} \int f(\Omega_N)\delta\left(\sum_{q=1}^{N}(\Omega_q - \bar{\omega}_q)\right) d\Omega_N \\ &= \frac{i\prod_{k=1}^{N}\mu_k}{(-2\pi)^{N-1}} \int f(\bar{\omega}_N - \sum_{q=1}^{N-1}(\Omega_q - \bar{\omega}_q)) \prod_{j=1}^{N-1} \frac{f(\Omega_j)d\Omega_j}{\sum_{p=1}^{j}(\Omega_p - \bar{\omega}_p)}. \end{aligned} \tag{15}$$

An important property arising from Eq. (15) is that for the state N to be populated by the pulse $E(t)$, the sum of the transition frequencies should be equal to $\varepsilon_N - \varepsilon_0$. There is an important difference with the work of Larsen and Bloembergen[34], in which the condition $\varepsilon_N - \varepsilon_0 = N\omega$ leads to multiphoton Rabi oscillations, not to an actual transition, as they have a stationary periodic field, not a radiation pulse.

We will consider the case where M, the number of the components in the pulse Eq. (2), is equal to N, the number of the transitions in the multilevel ladder system, so

$$E\left(t\right)=s\left(t\right)\sum_{l=1}^{N}A_{l}e^{-i\theta_{l}}e^{-i\omega_{l}t}+c.c, \tag{16a}$$

$$f(\omega)=\sum_{l=1}^{N}\left[A_{l}e^{-i\theta_{l}}S\left(\omega-\omega_{l}\right)+A_{l}e^{i\theta_{l}}S\left(\omega+\omega_{l}\right)\right], \tag{16b}$$

where

$$S\left(\omega\right)=\int_{-\infty}^{\infty}s\left(t\right)e^{i\omega t}dt, \tag{17}$$

and each component is only resonant with the corresponding system transition,

$$\left|\omega_{k}-\bar{\omega}_{k}\right|\ll\left|\omega_{k}-\bar{\omega}_{j\neq k}\right|,\ k,j=1,\cdots,N \tag{18}$$

with the function $S\left(\omega\right)$ assumed to be smooth, $\left|dS/d\omega\right|\ll\left|S\right|/\omega_{k}$. The latter condition means that the typical duration of the pulse significantly exceeds the transition periods $2\pi/\omega_{k}$, $k=1,\cdots,N$. Then the nearly resonant terms in Eq. (15) are kept,

$$C_{N}\approx\frac{i\prod_{k=1}^{N}\mu_{k}e^{-i\theta_{k}}}{\left(-2\pi\right)^{N-1}}\int A_{N}S(\bar{\omega}_{N}-\omega_{N}-\sum_{q=1}^{N-1}\left(\Omega_{q}-\bar{\omega}_{q}\right))\prod_{j=1}^{N-1}\frac{A_{j}S(\Omega_{j}-\omega_{j})d\Omega_{j}}{\sum_{p=1}^{j}\left(\Omega_{p}-\bar{\omega}_{p}\right)} \tag{19a}$$

$$=\tilde{C}_{N}\prod_{k=1}^{N}\mu_{k}A_{k}e^{-i\theta_{k}}, \tag{19b}$$

where

$$\tilde{C}_{N}=\frac{i}{\left(-2\pi\right)^{N-1}}\int S(\bar{\omega}_{N}-\omega_{N}-\sum_{q=1}^{N-1}\left(\Omega_{q}-\bar{\omega}_{q}\right))\prod_{j=1}^{N-1}\frac{S(\Omega_{j}-\omega_{j})d\Omega_{j}}{\sum_{p=1}^{j}\left(\Omega_{p}-\bar{\omega}_{p}\right)}. \tag{20}$$

If each component of the pulse is exactly resonant with a transition of the system,

$$\omega_{k}=\bar{\omega}_{k}, \tag{21}$$

then a simple form for $\tilde{C}_N$,

$$\tilde{C}_N = \frac{i^N \tau^N}{N!}, \tag{22}$$

may be attained with

$$\tau = S(0) = \int_{-\infty}^{\infty} s(t)\, dt \tag{23}$$

being the effective pulse duration. It's easier to prove Eq. (22) in the time domain than in the frequency domain. Considering the control field in Eq. (16a), whose each component is resonant with a particular transition, the neglect of all nonresonant terms in the Eq. (9) yields

$$C_N \approx i^N \prod_{k=1}^{N} \mu_k A_k e^{-i\theta_k} \int_{-\infty}^{\infty} dt_N s(t_N) \int_{-\infty}^{t_N} dt_{N-1} s(t_{N-1}) \ldots \int_{-\infty}^{t_2} dt_1 s(t_1) \tag{24a}$$

$$= i^N \prod_{k=1}^{N} \mu_k A_k e^{-i\theta_k} \frac{\left[\int_{-\infty}^{\infty} s(t)\, dt\right]^N}{N!} \tag{24b}$$

$$= \frac{i^N \tau^N}{N!} \prod_{k=1}^{N} \mu_k A_k e^{-i\theta_k}, \tag{24c}$$

from which Eq. (22) follows.

If the pulse is not exactly resonant

$$\delta_k = \omega_k - \bar{\omega}_k \neq 0, \tag{25}$$

then $\tilde{C}_N$ is not so simple, except for the case where $\delta_k = \delta$ is independent of k, i.e., the detuning is the same for all frequencies. In this case Eq. (22) still applies, but Eq. (23)

should be modified to become

$$\tau = S\left(-\delta\right) = \int_{-\infty}^{\infty} s\left(t\right) e^{-i\delta t} dt. \tag{26}$$

Introducing the following change of variable in Eq. (20),

$$z_j = \Omega_j - \omega_j, \tag{27}$$

yields

$$\tilde{C}_N = \frac{i}{\left(-2\pi\right)^{N-1}} \int S(-\sum\nolimits_{q=1}^{N-1} z_q - \Delta_N) \prod_{j=1}^{N-1} \frac{S(z_j) dz_j}{\sum_{p=1}^{j} z_p + \Delta_j}, \tag{28}$$

where Δ_k is the cumulant detuning,

$$\Delta_k = \sum_{p=1}^{k} \delta_p, k = 1, \cdots, N. \tag{29}$$

Inserting delta functions in Eq. (28) produces

$$\begin{aligned} \tilde{C}_N &= \frac{i}{\left(-2\pi\right)^{N-1}} \prod_{k=1}^{N-1} \left(\int_{-\infty}^{\infty} dz_k \int_{-\infty}^{\infty} dz_k' \right) S(-\sum\nolimits_{q=1}^{N-1} z_q - \Delta_N) \prod_{j=1}^{N-1} \frac{S(z_j)\delta\left(z_j - z_j'\right)}{z_j' + \sum_{p=1}^{j-1} z_p + \Delta_j} \\ &= \frac{\left(-1\right)^{N-1} i}{\left(2\pi\right)^{2(N-1)}} \prod_{k=1}^{N-1} \left(\int_{-\infty}^{\infty} dz_k \int_{-\infty}^{\infty} dz_k' \int_{-\infty}^{\infty} d\tau_k \right) S(-\sum_{q=1}^{N-1} z_q - \Delta_N) \prod_{j=1}^{N-1} S(z_j) e^{i\tau_j z_j} \frac{e^{-i\tau_j z_j'}}{z_j' + \sum_{p=1}^{j-1} z_p + \Delta_j}. \end{aligned} \tag{30}$$

From Eq. (13) it follows that there is a small positive imaginary part in each variable z_k' in Eq. (27), so the integration over z_k', $k = 1, \cdots, N-1$ yields

$$\tilde{C}_N = \frac{i^N}{\left(2\pi\right)^{N-1}} \prod_{k=1}^{N-1} \left(\int_{0}^{\infty} d\tau_k \int_{-\infty}^{\infty} dz_k \right) G(-\sum_{q=1}^{N-1} z_q - \Delta_N) \prod_{j=1}^{N-1} G(z_j) e^{i\tau_j \left(\sum_{p=1}^{j} z_p + \Delta_j\right)}. \tag{31}$$

The same result can also be obtained directly from Eq. (12) by changing from $t_1, \cdots, t_{N-1}$ to $\tau_1 = t_1, \tau_2 = t_2 - t_1, \cdots, \tau_N = t_N - t_{N-1}$ (or $t_k = \tau_1 + \cdots + \tau_k$, $k = 1, \cdots, N$) and from

Ω_j to z_j given by Eq. (27). As illustrations of the general formulation above, Gaussian and rectangular pulses are considered in the following treatment.

A. Gaussian pulse

If the pulse envelope of the control field is Gaussian,

$$s\left(t\right) = \exp\left(-\frac{\pi t^2}{\tau^2}\right), \tag{32a}$$

$$S\left(\omega\right) = \tau \exp\left(-\frac{\omega^2}{\sigma^2}\right), \tag{32b}$$

with σ being the spectral width of the pulse,

$$\sigma = \frac{2\sqrt{\pi}}{\tau}, \tag{33}$$

then substituting Eq. (32b) into Eq. (31) leads to

$$\begin{aligned} \tilde{C}_N &= \frac{i^N \tau^N}{\left(2\pi\right)^{N-1}} \prod_{k=1}^{N-1} \left(\int_0^\infty d\tau_k \int_{-\infty}^\infty dz_k\right) \exp\left[-\left(\sum_{q=1}^{N-1} z_q + \frac{\Delta_N}{\sigma}\right)^2 - \sum_{j=1}^{N-1} z_j^2 + \sum_{j=1}^{N-1} i\tau_j \left(\sum_{p=1}^{j} z_j + \frac{\Delta_j}{\sigma}\right)\right] \\ &= \frac{i^N \tau^N}{\left(2\pi\right)^{N-1}} \prod_{k=1}^{N-1} \left(\int_0^\infty d\tau_k \int_{-\infty}^\infty dz_k\right) \exp\left[-\sum_{k,j=1}^{N-1} z_k A_{kj} z_j + \sum_{j=1}^{N-1} b_j z_j + i \sum_{j=1}^{N-1} \tau_j \frac{\Delta_j}{\sigma} - \frac{\Delta_N^2}{\sigma^2}\right], \end{aligned} \tag{34}$$

where the elements of matrix A and vector b are

$$A_{kj} = \boldsymbol{\delta}_{kj} + 1, \tag{35a}$$

$$b_j = i \sum_{p=j}^{N-1} \tau_p - 2\frac{\Delta_N}{\sigma}. \tag{35b}$$

Carrying out the Gaussian integrals in Eq. (34) with respect to z_k, $k = 1, \cdots, N-1$ yields

$$\tilde{C}_N = \frac{i^N \tau^N}{(2\pi)^{N-1}} \sqrt{\frac{\pi^{N-1}}{\det A}} \prod_{k=1}^{N-1} \left(\int_0^\infty d\tau_k \right) \exp \left[\frac{1}{4} \sum_{k,j=1}^{N-1} b_k \left(A^{-1}\right)_{kj} b_j + i \sum_{j=1}^{N-1} \tau_j \frac{\Delta_j}{\sigma} - \frac{\Delta_N^2}{\sigma^2} \right]. \tag{36}$$

It's easy to verify that

$$\left(A^{-1}\right)_{kj} = \boldsymbol{\delta}_{kj} - \frac{1}{N}, \tag{37a}$$

$$\det A = N. \tag{37b}$$

Performing some algebra produces the perturbative solution for the Gaussian pulse,

$$\tilde{C}_N = \frac{i^N \tau^N}{2^{N-1} \pi^{\frac{N-1}{2}} N^{\frac{1}{2}}} e^{-\frac{\Delta_N^2}{N\sigma^2}} \prod_{p=1}^{N-1} \left[\int_0^\infty \exp\left(-\frac{i}{N\sigma} \tau_p D_p \right) d\tau_p \right] \exp\left\{ -\frac{1}{4N} P_N(\vec{\tau}) \right\}, \tag{38}$$

where

$$D_k = k\Delta_N - N\Delta_k,\ k = 1, \cdots, N-1, \tag{39a}$$

$$P_N(\vec{\tau}) = \sum_{k=1}^{N-1} k(N-k)\tau_k^2 + \sum_{0<k<j<N} 2k(N-j)\tau_k\tau_j. \tag{39b}$$

When $D_k \neq 0$ and $\sigma \to 0$, the asymptotic behavior of transition probability[37],

$$|C_N| \propto \sigma^{N-1} \frac{\exp\left(-\frac{\Delta_N^2}{N\sigma^2}\right)}{\prod_{k=1}^{N-1} |D_k|}, \tag{40}$$

is obtained. The above equation shows that the yield exponentially decays with the sum of the detuning of the individual transitions, Δ_N from Eq. (29). This behavior can be easily understood since, as pointed out previously, the sum of the field frequencies should be equal to the sum of the transition frequencies of the system, which is the energy conservation

condition. A simple calculation shows that, for all $|\delta_i| \gg \sigma$, the most probable way of meeting the energy conservation constraint leads to $\left|\tilde{C}_N\right| \propto \exp\left(-\Delta_N^2/N\sigma^2\right)$.

B. Rectangular pulse

In the case that the pulse envelope of the control field is rectangular,

$$s(t) = \begin{cases} 1, & 0 < t < T, \\ 0, & \text{otherwise.} \end{cases} \tag{41a}$$

$$S(\omega) = \frac{e^{i\omega T} - 1}{i\omega}, \tag{41b}$$

the substitution of Eq. (41b) into Eq. (28) leads to

$$\tilde{C}_N = \frac{i}{(-2\pi)^{N-1}} \int \mathcal{I}_{N-1} \prod_{j=1}^{N-2} \frac{e^{iz_jT} - 1}{iz_j} \frac{dz_k}{\sum_{p=1}^{j} z_p + \Delta_j}, \tag{42}$$

where $\mathcal{I}_{N-1}$ is an integral with respect to z_{N-1}

$$\begin{aligned} \mathcal{I}_{N-1} &= \int \frac{\exp\left[-i\left(\sum_{q=1}^{N-1} z_q + \Delta_N\right)T\right] - 1}{-i\left[\sum_{q=1}^{N-1} z_q + \Delta_N\right]} \frac{e^{iz_{N-1}T} - 1}{iz_{N-1}} \frac{dz_{N-1}}{\sum_{p=1}^{N-1} z_p + \Delta_{N-1}} \\ &= \int \frac{\exp\left[-i\left(\sum_{q=1}^{N-1} z_q + \Delta_N\right)T\right]}{-i\left[\sum_{q=1}^{N-1} z_q + \Delta_N\right]} \frac{-1}{iz_{N-1}} \frac{dz_{N-1}}{\sum_{p=1}^{N-1} z_p + \Delta_{N-1}}. \end{aligned} \tag{43}$$

This equation can be evaluated by the residue theorem[37]. After checking the three poles in the lower half plane $\operatorname{Im} z_{N-1} \leq 0$, it can be shown that only the residue of the zero pole is needed because those of the other two poles do not contribute after integrating with respect

to z_{N-2}, so we have

$$\begin{aligned}\mathcal{I}_{N-1} &= -2\pi i \mathrm{Res}\left(z_{N-1}=0\right) \\ &= \frac{2\pi i}{\sum_{q=1}^{N-2} z_q + \Delta_{N-1}} \frac{\exp\left[-i\left(\sum_{q=1}^{N-2} z_q + \Delta_N\right)T\right]}{\sum_{q=1}^{N-2} z_q + \Delta_N}.\end{aligned} \tag{44}$$

Integrating with respect to z_{N-2}, z_{N-3},$\cdots$, z_2, similarly as above, reduces C_N to an integral with respect to only one variable,

$$\begin{aligned}\tilde{C}_N &= \frac{(-1)^N i}{2\pi} \int \frac{e^{-i(z_1+\Delta_N)T}}{\prod_{q=2}^{N}(z_1+\Delta_q)} \frac{e^{iz_1T}-1}{iz_1} \frac{dz_1}{z_1+\Delta_1} \\ &= \frac{(-1)^N i}{2\pi} \int \frac{e^{-i(z_1+\Delta_N)T}}{\prod_{q=2}^{N}(z_1+\Delta_q)} \frac{-1}{iz_1} \frac{dz_1}{z_1+\Delta_1}.\end{aligned} \tag{45}$$

This result finally produces a compact form for the perturbative solution with a rectangular pulse:

$$\tilde{C}_N = \frac{(-1)^N i}{2\pi} \int \frac{e^{-i(z+\Delta_N)T}}{z\prod_{q=1}^{N}(z+\Delta_q)} dz. \tag{46}$$

If all of the transitions are resonant: $\delta_p = 0$, $p = 1, \cdots, N$, then

$$\begin{aligned}\tilde{C}_N &\approx \frac{(-1)^N i}{2\pi} \int \frac{e^{-izT}}{z^{N+1}} dz \\ &= \frac{i^N}{N!} T^N,\end{aligned} \tag{47}$$

which is consistent with Eq. (22). If all Δ_q, $q = 1, \cdots, N$, are different from each other, then from Eq. (46),

$$\tilde{C}_N = (-1)^N \sum_{q=0}^{N} e^{-i(\Delta_N-\Delta_q)T} \prod_{j=0(j\neq q)}^{N} (\Delta_j - \Delta_q)^{-1} \tag{48}$$

with $\Delta_0 = 0$. In the case where all of the detunings are the same, we have $\Delta_q = q\delta$, then

$$\tilde{C}_N = \frac{(-1)^N}{N!}\delta^{-N}\left(e^{-iT\delta} - 1\right)^N. \tag{49}$$

This expression agrees with Eq. (22) derived earlier for a pulse of an arbitrary shape.

An interesting consequence of Eq. (49) is that the transition probability is an oscillating function of $T\delta$,

$$\left|\tilde{C}_N\right|^2 = \frac{2^{2N}}{(N!)^2}\delta^{-2N}\sin^{2N}\frac{T\delta}{2}. \tag{50}$$

In particular, for $T\delta = 2n\pi$, $n = 0, 1, \cdots$, $\left|\tilde{C}_N\right|^2$ becomes zero. Such an antiresonance is a result of destructive interference, which eliminates transitions to higher states.

IV. COOPERATION BETWEEN A WEAK FIELD AND NOISE FOR A MULTI-STATE LADDER SYSTEM

From Eq. (19b), it is evident that, in a weak field driven multilevel ladder system, the transition probability is independent of the phases of the individual pulse components, but is sensitive to their amplitudes and frequencies. This section considers the noise-averaged transition probability over the amplitudes and frequencies of a weak field, and shows that the population transfer can be enhanced under suitable conditions.

A. Noise in the amplitudes of the control field

From Eq. (19b), it is easy to determine the dependence of the control yield on the amplitudes of the field pulses,

$$O\left[E(t)\right]=\left|C_N\right|^2\approx\alpha_N^2\prod_{l=1}^{N}A_l^2, \tag{51}$$

where

$$\alpha_N=\left|\tilde{C}_N\prod_{k=1}^{N}\mu_k\right| \tag{52}$$

is independent of the amplitudes. If each amplitude A_l is contaminated with random uniform noise distributed over $[-\gamma_l,\gamma_l]$, as in Eq. (3a), then the noise-averaged outcome of the control process is

$$\bar{O}\left[E(t),\vec{\gamma}\right]=\alpha_N^2\prod_{l=1}^{N}\left\langle A_l^2\right\rangle, \tag{53a}$$

where $\langle A_l^2\rangle$ is the mean square amplitude of the l-th radiation component,

$$\left\langle A_l^2\right\rangle=\int(A_l^0+\tilde{x}_l)^2\rho_l^{(A)}\left(\tilde{x}_l\right)d\tilde{x}_l={A_l^0}^2+\left\langle\tilde{x}_l^2\right\rangle. \tag{54}$$

It is instructive to compare this expression with the outcome from the control field not having amplitude fluctuations,

$$O^{(0)}\left[E\left(t\right)\right]=\alpha_N^2\prod_{l=1}^{N}{A_l^0}^2. \tag{55}$$

The ratio

$$\bar{O}/O^{(0)} = \prod_{l=0}^{N} \frac{\langle A_l^2 \rangle}{{A_l^0}^2} \tag{56}$$

becomes appreciable for a large number of states N, even when the ratio $\langle A_l^2 \rangle \, / \, {A_l^0}^2$ is close to 1.0 for each individual transition. For example, if $\langle A_l^2 \rangle \, / \, {A_l^0}^2 = 1 + \varepsilon$ with $\varepsilon \ll 1$, then $\bar{O}/O^{(0)} \cong 1 + N\varepsilon$.

To find the optimal field amplitude, we set $\langle A_l^2 \rangle = {A_l^0}^2 + \langle \tilde{x}_l^2 \rangle$ and minimize the functional

$$J_N\left(A_n^{(0)}\right) = \left| \alpha_N^2 \prod_{l=1}^{N} ({A_l^0}^2 + \left\langle \tilde{x}_l^2 \right\rangle) - O_T \right|^2 + \alpha \sum_{l=0}^{N} {A_l^0}^2 \tag{57}$$

over A_l^0. Assuming that $\{\langle \tilde{x}_l^2 \rangle\}$ are independent of $\{A_l^0\}$, the optimal value of A_l^0 satisfies

$${A_l^0}^2 + \left\langle \tilde{x}_l^2 \right\rangle = \text{Constant}, \tag{58}$$

independent of l. An important consequence is that, for given $\{\langle \tilde{x}_l^2 \rangle\}$, the contribution from noise can beneficially serve to decrease the required amplitude A_l^0 of the optimal control field leading to a given yield, provided that the yield is small. Cooperation with noise can be extended to modest control yields beyond the perturbation approximation, as shown in numerical simulations[11]. Although the presence of strong noise can considerably reduce the coherent nature of the dynamics, modest target yields can still be reached, including in a very efficient manner. However, when attempting to achieve high control yields, a different mechanism is generally operative involving competition between the deterministic portion of the control field and the noise[11].

B. Noise in the control frequency spectrum

Here we consider a weak control field with a Gaussian envelope [Eq. (32)] and frequency noise having Gaussian distribution

$$f_k\left(\delta_k\right) = \frac{1}{d_k\sigma\sqrt{\pi}} \exp\left[-\left(\frac{\delta_k - \bar{\delta}_k}{d_k\sigma}\right)^2\right], \tag{59}$$

where the random variables are the noise contaminated detunings

$$\delta_k = \bar{\delta}_k + \tilde{z}_k = \omega_k^0 + \tilde{z}_k - \bar{\omega}_k \tag{60}$$

with $\{\tilde{z}_k\}$ being uncertainties in the laser frequencies $\{\omega_k\}$ in Eq. (3c). An additional source of the detuning noise can be from the transition frequencies $\{\bar{\omega}_k\}$ due to the Doppler or phonon-induced shift in the control medium. Recalling Eq. (38) yields the noise-averaged transition probability,

$$\begin{aligned}\left\langle |C_N|^2 \right\rangle &= \prod_{q=1}^{N} \left(\int_{-\infty}^{\infty} f_q\left(\delta_q\right) d\delta_q\right) |C_N|^2 \\ &= \frac{\tau^{2N}}{\left(4\pi\right)^{N-1} N} \prod_{p=1}^{N-1} \left(\int_0^{\infty} d\tau_p \int_0^{\infty} d\tau_p'\right) L_N\left(\vec{\tau}, \vec{\tau}'\right) \exp\left\{-\frac{1}{4N}\left[P_N\left(\vec{\tau}\right) + P_N\left(\vec{\tau}'\right)\right]\right\}.\end{aligned} \tag{61}$$

Here, L_N is

$$\begin{aligned}L_N\left(\vec{\tau}, \vec{\tau}'\right) &= \prod_{q=1}^{N} \left(\int_{-\infty}^{\infty} f_q\left(\delta_q\right) d\delta_q\right) \exp\left[-\frac{2}{N\sigma^2}\Delta_N^2 - \frac{i}{N\sigma}\sum_{k=1}^{N-1}\left(\tau_k - \tau_k'\right) D_k\right] \\ &= \frac{1}{\sigma^N \pi^{N/2} \prod_{k=1}^{N} d_k} \prod_{q=1}^{N} \left(\int_{-\infty}^{\infty} d\delta_q\right) \exp\left[-\frac{1}{\sigma^2}\sum_{k,j=1}^{N} \delta_k B_{kj} \delta_j + \sum_{k=1}^{N} \delta_k c_k - \sum_{k=1}^{N} \frac{\bar{\delta}_k^2}{d_k^2 \sigma^2}\right]\end{aligned} \tag{62}$$

with the elements of the matrix B and the vector c being

$$B_{kj} = \frac{1}{d_k^2}\boldsymbol{\delta}_{kj} + \frac{2}{N}, \tag{63a}$$

$$c_k = \frac{2\bar{\delta}_k}{\sigma^2 d_k^2} - \frac{i}{\sigma}\left(\bar{V} - V_k\right), \tag{63b}$$

and the parameters V_k and $\vec{V}$ specified by

$$V_k = \sum_{j=k}^{N-1}\left(\tau_j - \tau_j'\right),\ k = 0, \cdots, N-1;\ V_N = 0, \tag{64a}$$

$$\bar{V} = \frac{1}{N}\sum_{k=1}^{N} V_k. \tag{64b}$$

Integrating Eq. (62) with respect to δ_q yields

$$L_N\left(\vec{\tau}, \vec{\tau}'\right) = \frac{1}{\prod_{k=1}^{N} d_k}\sqrt{\frac{1}{\det B}}\exp\left[\frac{\sigma^2}{4}\sum_{k,j=1}^{N} c_k\left(B^{-1}\right)_{kj} c_j - \sum_{k=1}^{N}\frac{\bar{\delta}_k^2}{d_k^2\sigma^2}\right], \tag{65}$$

with the elements of B^{-1} (inverse matrix of B) and the determinant of B being

$$\left(B^{-1}\right)_{kj} = d_k^2\left(\boldsymbol{\delta}_{kj} - \frac{2d_j^2}{N\left(1 + 2\bar{d}^2\right)}\right), \tag{66a}$$

$$\det B = \frac{1 + 2\bar{d}^2}{\prod_{k=1}^{N} d_k^2},\ \bar{d}^2 = \frac{1}{N}\sum_{k=1}^{N} d_k^2. \tag{66b}$$

The expression for the scaled transition probability is simplified in two limiting cases. The first case is that of nearly resonant driving, where $\left|\bar{\delta}_k\right| \ll \sigma d_k$. In this case $c_k = -i\sigma^{-1}\left(\bar{V} - V_k\right)$ and the term proportional to $\bar{\delta}_k^2$ in Eq. (65), can be neglected. However, it is easy to see that $L_N\left(\vec{\tau}, \vec{\tau}'\right) < 1$ because of the noise in the frequency spectrum, and L_N rapidly decreases with the increasing noise intensity parameters d_k. Therefore, the

transition probabilities $\langle |C_N|^2 \rangle$ with noise are smaller than without noise when the control field frequencies are reliably tuned to be resonant with the system transition frequencies.

The role of the noise is reversed in the case of comparatively strong detuning, $|\bar{\delta}_k| \gg \sigma d_k$. In this case we have $c_k \approx 2\bar{\delta}_k/\sigma^2 d_k^2$, then Eqs. (61) and (65) produce

$$\langle |C_N|^2 \rangle \propto \exp\left[-\frac{4\bar{\Delta}_N^2}{N\sigma^2\left(1+2\bar{d}^2\right)}\right] \tag{67}$$

with $\bar{\Delta}_N = \sum_{k=1}^{N} \bar{\delta}_k$. In this region, it follows from Eq. (67) that increasing the noise intensity parameter $\bar{d}^2$ leads to an exponentially strong increase in the transition probability. This finding shows how noise can play a constructive role in a controlled quantum system. This result may be understood from the fact that for strong detuning, spectral noise can lead to some pulses actually being closer to resonance. For these pulses the transition probability is exponentially higher than for nonresonant pulses. As a result, the noise averaged transition probability is also strongly increased.

Noise-induced enhancement of the transition probability occurs also for rectangular laser pulses. This is most easy to see when the detunings of all the frequency components in the pulse are the same and the scaled transition probability is given by Eq. (50). As noted earlier, the transition amplitude into the target state is completely eliminated if the detuning satisfies $\delta = 2n\pi/T$. Pulse-to-pulse variation of δ, or pulse-to-pulse variation of the duration T will suppress the antiresonance and lead to a nonzero transition probability even when $\left|\tilde{C}_N\right|^2 = 0$ for the average values $\delta = \bar{\delta}$, $T = \bar{T}$. If the width of the distribution over δ is small

compared to $\bar{\delta}$, but largely exceeds π/T, then from Eq. (50) we have $\left\langle \left|\tilde{C}_N\right|^2 \right\rangle \approx \frac{(2N)!}{(N!)^4}\bar{\delta}^{-2N}$.

V. CONCLUSIONS

This paper explores the dynamics of population transfer in a multistate ladder quantum system driven by noisy control pulses, with particular emphasis on identifying circumstances when cooperation between the field and noise may occur. The noise enters as run-to-run uncertainties in the control amplitudes, phases and frequencies with the observation being an ensemble average over many runs as is commonly done in the laboratory. If the rotating wave approximation is valid, the quantum dynamics in the weak field limit is greatly simplified and independent of the control phases. Furthermore, if the objective yield is modest, the control field can cooperate with amplitude noise to reduce the applied fluence. Frequency noise in the control field is shown to be capable of enhancing the transition probability when the detuning is large. In the laboratory implementation of closed-loop control, the optimal field will be deduced to extract as much beneficial value as possible from the presence of noise. This paper presents a theoretical foundation showing that ample opportunity exists to take advantage of noise.

The above conclusions are fully consistent with recent numerical simulations[11]. Although the analytical treatment in this paper only applies to ladder-configuration systems,

the basic conclusions on the prospects for cooperating with noise should be applicable for optimal control of many multistate systems. This point is confirmed with a non-ladder system[11] where a high degree of cooperation was found between the noise and the deterministic part of the field. Regardless of whether it is dynamically beneficial to fight or cooperate with noise, the optimal field will appropriately emphasize the dynamical pathways that correspondingly either work with or circumvent the influence of the noise[11].

Acknowledgments

The authors acknowledge support from the Department of Energy and NSF-PHY 0555346.

[1] S. A. Rice and M. Zhao, *Optical Control of Molecular Dynamics* (Wiley, New York, 2000).

[2] H. Rabitz, M. Hsieh, and C. Rosenthal, Science **303**, 1998 (2004).

[3] T. Brixner, N. H. Damrauer, and G. Gerber, in *Advances in Atomic, Molecular, and Optical Physics*, edited by B. Bederson and H. Walther (Academic, San Diego, CA, 2001), vol. 46, pp. 1–54.

[4] I. Walmsley and H. Rabitz, Phys. Today **56**, 43 (2003).

[5] A. I. Burshtein, A. A. Zharikov, and V. S. Malinovsky, Phys. Rev. A **43**, 1538 (1991).

[6] R. Walser, H. Ritsch, P. Zoller, and J. Cooper, Phys. Rev. A **45**, 468 (1992).

[7] O. E. Akramine, A. Makhoute, M. Zitane, and M. Tij, Phys. Rev. A **58**, 4892 (1998).

[8] J. M. Geremia, W. Zhu, and H. Rabitz, J. Chem. Phys. **113**, 10841 (2000).

[9] H. Rabitz, Phys. Rev. A **66**, 63405 (2002).

[10] I. R. Sola and H. Rabitz, J. Chem. Phys. **120**, 9009 (2004).

[11] F. Shuang and H. Rabitz, J. Chem. Phys. **121**, 9270 (2004).

[12] F. Grossmann, T. Dittrich, P. Jung, and P. Hänggi, J. Stat. Phys. **70**, 229 (1993).

[13] P. Blanchard, G. Bolz, M. Cini, G. F. Deangelis, and M. Serva, J. Stat. Phys. **75**, 749 (1994).

[14] J. S. Shao, C. Zerbe, and P. Hänggi, Chem. Phys. **235**, 81 (1998).

[15] B. G. Klappauf, W. H. Oskay, D. A. Steck, and M. G. Raizen, Phys. Rev. Lett. **81**, 1203 (1998).

[16] P. Gross, D. Neuhauser, and H. Rabitz, J. Chem. Phys. **98**, 4557 (1992).

[17] G. J. Tóth, A. Lőrincz, and H. Rabitz, J. Chem. Phys. **101**, 3715 (1994).

[18] S. Rana, L. D. Whitley, and R. Cogswell, in *Proceedings of the Fourth International Conference on Parallel Problem Solving from Nature, Berlin, Germany, September 1996*, edited by H. Voigt, W. Ebeling, I. Rechenberg, and H.-P. Schwefel (Springer, Berlin, 1996), pp. 198–207.

[19] A. Assion, T. Baumert, M. Bergt, T. Brixner, B. Kiefer, V. Seyfried, M. Strehle, and G. Gerber, Science **282**, 919 (1998).

[20] J. Kunde, B. Baumann, S. Arlt, F. Morier-Genoud, U. Siegner, and U. Keller, Appl. Phys.

Lett. **77**, 924 (2000).

[21] D. Zeidler, S. Frey, K.-L. Kompa, and M. Motzkus, Phys. Rev. A **64**, 23420 (2001).

[22] T. Brixner, N. H. Damrauer, P. Niklaus, and G. Gerber, Nature (London) **414**, 57 (2001).

[23] R. J. Levis, G. M. Menkir, and H. Rabitz, Science **292**, 709 (2001).

[24] C. Daniel, J. Full, L. González, C. Lupulescu, J. Manz, A. Merli, Š. Vajda, and L. Wöste, Science **299**, 536 (2003).

[25] R. S. Judson and H. Rabitz, Phys. Rev. Lett. **68**, 1500 (1992).

[26] M. Dykman, D. Luchinsky, R. Mannella, P. McClintock, N. Stein, and N. Stocks, Nuov. Cim. D **17**, 661 (1995).

[27] K. Wiesenfeld and F. Jaramillo, CHAOS **8**, 539 (1998).

[28] F. Shuang, C. Yang, H. Zhang, and Y. Yan, Phys. Rev. E **61**, 7192 (2000).

[29] V. N. Smelyanskiy and M. I. Dykman, Phys. Rev. E **55**, 2516 (1997).

[30] B. E. Vugmeister and H. Rabitz, Phys. Rev. E **55**, 2522 (1997).

[31] A. Lindinger, S. M. Weber, C. Lupulescu, F. Vetter, M. Plewicki, A. Merli, L. Woste, A. F. Bartelt, and H. Rabitz, Phys. Rev. A **71**, 013419 (2005).

[32] K.-J. Boller, A. Imamolu, and S. E. Harris, Phys. Rev. Lett. **66**, 2593 (1991).

[33] S. Balle, N. B. Abraham, P. Colet, and M. S. Miguel, IEEE J. Quantum Electron. **29**, 33 (1993).

[34] D. M. Larsen and N. Bloembergen, Opt. Commu. **17**, 254 (1976).

[35] A. Wallraff, D. Schuster, A. Blais, L. Frunzio, R. Huang, J. Majer, S. Kumar, S. Girvin, and R. S. R. Schoelkopf, Nature (London) **431**, 162 (2004).

[36] M. I. Dykman and M. V. Fistul, Phys. Rev. B **71**, 140508(R) (2005).

[37] P. Henrici, *Applied And Computational Complex Analysis*, vol. 2 (John Wiley and Sons, New York, 1977).